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THE EFFECTS OF IRRADIATION ON URANIUM-PLUTONIUM-FISSIUM FUEL ALLOYS

by

J. A. Horak, J. H. Kittel,
and R. J. Dunworth

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THE EFFECTS OF IRRADIATION ON URANIUM-PLUTONIUM-FISSIUM FUEL ALLOYS

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ABSTRACT

A total of 35 specimens of uranium-plutonium-fissium alloy and 2 specimens of uranium-10 w/o plutonium-5 w/o molybdenum alloy were irradiated as a part of the fuel-alloy development program for fast breeder reactors at Argonne National Laboratory. Total atom burnups ranged from 1.0 to 1.8 per cent at maximum fuel temperatures ranging from 230 to 470°C. Emphasis was placed on the EBR-II Core-II reference fuel material, which is an injection-cast, uranium-20 w/o plutonium-10 w/o fissium alloy. It was found that this material begins to swell catastrophically at irradiation temperatures above 370°C. The ability of the fuel to resist swelling did not appear to vary appreciably with minor changes in zirconium or fissium content. Decreasing the plutonium to 10 w/o, however, significantly improved the swelling behavior of the alloy. Both pour-cast and thermally cycled material and pour-cast, extruded, and thermally cycled material appeared to be more stable under irradiation than injection-cast material. Under comparable irradiation conditions, the specimens of uranium-20 w/o plutonium-5 w/o molybdenum alloy were less dimensionally stable than the uranium-plutonium-fissium alloys investigated.

INTRODUCTION

The fast power breeder reactor is an important part of the United States Atomic Energy Program for the development of advanced civilian power reactors. The Experimental Breeder Reactor Number II (EBR-II) has been built to demonstrate the breeder concept and the pyrometallurgical reprocessing of spent fuel in a completely integrated fuel cycle.

In the reprocessing techniques developed at Argonne National Laboratory for fuel elements discharged from fast breeder reactors, a group of metallic fission product elements are not completely removed from the fuel. After several fuel irradiations and recycles, these elements therefore build

up to an equilibrium composition. If the reprocessing procedures and efficiencies remain constant, the equilibrium ratio of these elements among themselves is a function of the burnup to which the fuel is taken. This group of fission products, which are elements of the second long period, has been named "fissium" and symbolized as Fs. Because of differences in fission yields, the amount and composition of the fissium which accumulates in the fuel depends upon whether uranium-235 or plutonium-239 is used as the fuel.

The fuel for the first core of EBR-II will be an alloy of uranium and fissium. With uranium-235 as the fissionable species, the reactor would operate only as a converter and not a breeder; therefore, subsequent fuel loadings will contain plutonium-239 as the fissionable species, which will qualify the reactor as a power-breeder reactor. In addition, the utilization of plutonium-239 as the fissionable species rather than uranium-235 increases the theoretical breeding gain from slightly under 1.2 to at least 1.5 for this reactor.[1]

The fuel for the second core of EBR-II will be an alloy of uranium-plutonium-fissium. The composition of the fissium addition is given in the following section. A series of irradiations have been made to determine the irradiation behavior of several uranium-plutonium-fissium alloys. This report presents the information obtained by the irradiation of 35 specimens of uranium-plutonium-fissium alloy and 2 specimens of uranium-plutonium-molybdenum. Preliminary irradiation results obtained with some of the alloys have been reported elsewhere.[2,3]

SPECIMEN MATERIAL AND PREPARATION OF IRRADIATION SPECIMENS

The materials from which the irradiation specimens were produced were reactor-grade normal uranium, production-grade plutonium, and a master alloy of uranium and the fissium elements. Table I gives a typical composition of the fissium mixture used in this investigation. This composition simulates an alloy containing the anticipated equilibrium amount of fission products as the result of several EBR-II irradiation and reprocessing cycles of a fuel in which plutonium-239 is the fissionable species. In the synthetic alloy, molybdenum and ruthenium have been substituted for technetium, which is produced only during fission. A typical composition of reconstituted fuel which will exist after several cycles of irradiation of the uranium-20 w/o plutonium-10 w/o fissium fuel is shown in Table II. In order to feed to the EBR-II reprocessing plant spent fuel which is not constantly changing in composition during early stages of reactor operation, it has been decided to fuel the reactor at the start with the synthetic fissium alloy shown in Table I.

Table I

COMPOSITION OF SYNTHETIC FISSIUM
USED IN IRRADIATION SPECIMENS

Element	w/o	Element	w/o
Zirconium	4.6	Rhodium	6.5
Molybdenum	25.9	Palladium	23.2
Ruthenium	39.8		

Table II

TYPICAL COMPOSITION OF IRRADIATED EBR-II
URANIUM-PLUTONIUM-FISSIUM FUEL ALLOY

Element	w/o	Element	w/o
Uranium	70.0	Technetium	0.7
Plutonium	20.0	Ruthenium	2.6
Zirconium	2.8	Rhodium	0.5
Molybdenum	2.4	Palladium	1.0

The alloy 70 w/o uranium-20 w/o plutonium-10 w/o fissium has been chosen as the reference fuel for the second core of EBR-II; 20 of the 37 specimens irradiated were of this composition. In an attempt to evaluate the effects of more or less plutonium on the irradiation stability of the reference fuel, 2 specimens containing 25 w/o plutonium-10 w/o fissium and 2 containing 10 w/o plutonium-10 w/o fissium were also irradiated. To study the effects of fissium concentrations higher or lower than that of the reference fuel, 2 specimens containing 20 w/o plutonium-15 w/o fissium and 2 containing 20 w/o plutonium-5 w/o fissium were irradiated. In addition, since molybdenum has been shown to be a beneficial alloying element in other uranium-base fuel alloys, 3 specimens of the composition uranium-20 w/o plutonium-5.4 w/o fissium-3.6 w/o molybdenum and 2 specimens of the composition uranium-20 w/o plutonium-5 w/o molybdenum were irradiated.

The reprocessing operation is less difficult if the fission product zirconium is not removed, but is allowed to build up to approximately one-half of the total normal fissium content. Therefore, 4 specimens containing the nominal fissium to zirconium ratio of 2:1 were irradiated to evaluate the effects of the additional zirconium on the irradiation behavior of the reference uranium-20 w/o plutonium-10 w/o fissium fuel alloy.

The specimens were fabricated by 3 methods. In the reprocessing operation the fuel pins will be fabricated by injection casting; 27 of the present specimens were therefore injection cast. The alloy was made by induction heating under vacuum in a graphite crucible which had been coated with a magnesium-zirconate and thoria wash. The melts were injection cast into precision-bore Vycor tubes which were 15.5 in. long by 0.145 in. ID; the Vycor tubes were internally coated with an Aquadag-thoria wash. The casting temperatures ranged from 1135 to 1230°C. The Vycor molds used were identical with those used in the production of the EBR-II, Core-I fuel. The purpose of this was to duplicate the casting conditions of the reprocessing facility as closely as possible, thereby minimizing the variables between the experimental specimens and the production fuel.

Two additional fabrication techniques were employed to study the effects of fabrication history on the irradiation behavior of these alloys. The other 10 specimens were induction melted under vacuum in high-purity thoria crucibles. With the exception of the uranium-20 w/o plutonium-5 w/o molybdenum composition, the melts were then top poured into graphite molds which were designed to give an extrusion billet large enough to give 2 extruded irradiation specimens and one as-cast irradiation specimen. The melts were poured at temperatures between 1450 and 1500°C. The billets were extruded bare in a heated die at temperatures between 390 and 590°C at a reduction of 7 to 1. The uranium-20 w/o plutonium-5 w/o molybdenum melt was top poured into copper molds of the same design as the previously mentioned graphite molds.

Seven of the irradiation specimens were thermally cycled prior to irradiation in order to determine the dimensional stability of the specimens under repeated temperature changes. The specimens were thermally cycled as follows: They were transferred mechanically from a sodium bath held at 50°C to one held at 500°C within a transfer time of one minute, held at 500°C for 5 min, returned to the bath at 50°C within one minute, and held at 50°C for 5 min. This cycle was repeated for a total of 200 cycles.

Figure 1 shows the typical preirradiation appearance of the specimens. The nominal preirradiation dimensions of the specimens were 1.00 in. long and 0.144 in. in diameter, which is also the diameter of the EBR-II fuel pins. The specimens for irradiation test were manufactured to the same diameters as the EBR-II fuel pins to eliminate effects due to cross-sectional size, such as radial temperature gradients and flux perturbation, when comparing the test specimens with the actual reactor fuel.

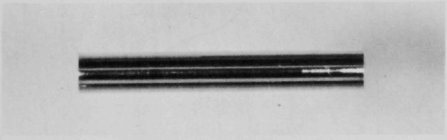

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Figure 1. Appearance of typical fuel specimen prior to irradiation.

EXPERIMENTAL PROCEDURE

The following properties of the specimens were measured prior to irradiation:

1. dimensions to 0.0001 in.;
2. weights to 0.1 mg;
3. density to 0.05 per cent by immersion in CCl_4; and
4. mass-spectrographic analysis for plutonium isotope concentration.

The specimens were irradiated at the Materials Testing Reactor (MTR) in 2 different groups, designated "EA" and "ANL-36." The capsules containing the EA specimens contained one specimen in each capsule, as shown in Figure 2. The resulting cylindrical geometry enabled the temperature gradients in the fuel and capsule to be calculated analytically. The capsules containing the ANL-36 specimens each contained 3 specimens located 120° apart, as shown in Figure 3. The temperature drops in these specimens and capsules were determined with the aid of an electrical geometrical analogue.(4) All of the capsules contained sufficient eutectic NaK to cover the specimens and provide adequate heat transfer to the process coolant water. The absolute gas pressure which existed over the NaK in these capsules after filling was approximately 1 x 10^{-4} mm of Hg.

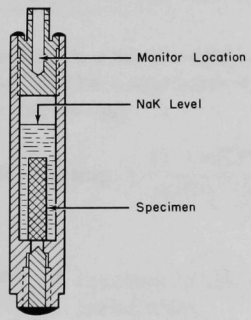

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Figure 2

Assembled irradiation capsule used for EA specimens. The capsule is 2-1/2 in. long and 1/2 in. in diameter

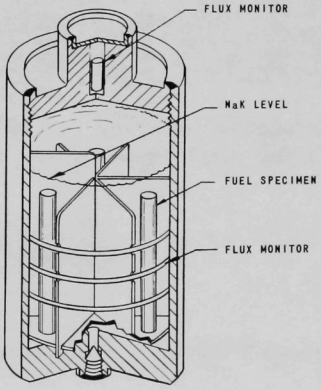

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Figure 3

Assembled irradiation capsule used for ANL-36 specimens. The capsule is 3-1/8 in. long and 1-1/8 in. in diameter

Each capsule contained an aluminum-0.5 w/o cobalt-0.5 w/o manganese monitor wire for neutron dosimetry, which was located at the top of the capsule as indicated in Figures 2 and 3. In addition to this monitor, the ANL-36 capsules contained an additional flux monitor wire which encircled the specimen holder at the longitudinal center of the specimens, as shown in Figure 3. The composition of this flux monitor was nickel-0.093 w/o cobalt. After corrections for resonance activation of the cobalt were made, the integrated neutron fluxes indicated by the dosimeters were used in computing preliminary temperature and burnup data on the specimens. Mass-spectrographic analyses for the plutonium-isotope composition were also performed on 8 of the 30 ANL-36 specimens and on 6 of the 7 EA specimens before and after irradiation. The preirradiation and post irradiation plutonium-isotope compositions of these 8 ANL-36 specimens are given in Table III. In addition to providing the most accurate measurements of burnup and temperature, these specimens also provided a calibration for the dosimeter-indicated burnup and temperature for the other specimens irradiated in the experiment.

Table III

PLUTONIUM ISOTOPE COMPOSITION OF ANL-36 SPECIMENS ANALYZED MASS SPECTROGRAPHICALLY BEFORE AND AFTER IRRADIATION

Specimen No.	Preirradiation Plutonium Composition, Mole Per Cent				Postirradiation Plutonium Composition, Mole Per Cent			
	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}
ANL-36-2-2	94.31 ± 0.05	5.27 ± 0.05	0.402 ± 0.004	0.0124 ± 0.0003	91.85 ± 0.08	7.66 ± 0.08	0.476 ± 0.008	0.0051 ± 0.0005
ANL-36-2-3	94.31 ± 0.05	5.27 ± 0.05	0.402 ± 0.004	0.0124 ± 0.0003	92.15 ± 0.06	7.37 ± 0.06	0.456 ± 0.007	0.023 ± 0.001
ANL-36-3-1	94.31 ± 0.05	5.27 ± 0.05	0.402 ± 0.004	0.0124 ± 0.0003	88.25 ± 0.11	11.34 ± 0.11	0.379 ± 0.008	0.0273 ± 0.0009
ANL 36-4-3	95.00 ± 0.05	4.71 ± 0.05	0.29 ± 0.01	Not Analyzed	91.33 ± 0.12	8.19 ± 0.12	0.451 ± 0.008	0.0235 ± 0.0011
ANL-36-6-3	94.31 ± 0.05	5.27 ± 0.05	0.402 ± 0.004	0.0124 ± 0.0003	90.32 ± 0.05	9.06 ± 0.05	0.584 ± 0.01	0.035 ± 0.002
ANL-36-7-1	94.31 ± 0.05	5.27 ± 0.05	0.402 ± 0.004	0.0124 ± 0.0003	91.17 ± 0.09	8.27 ± 0.09	0.530 ± 0.008	0.028 ± 0.002
ANL-36-9-3	94.31 ± 0.05	5.27 ± 0.05	0.402 ± 0.004	0.0124 ± 0.0003	89.61 ± 0.09	9.68 ± 0.09	0.674 ± 0.015	0.0424 ± 0.0013
ANL-36-10-3	94.31 ± 0.05	5.27 ± 0.05	0.402 ± 0.004	0.0124 ± 0.0003	89.32 ± 0.09	9.94 ± 0.09	0.691 ± 0.017	0.047 ± 0.006

The fuel burnups were computed by the first method presented in IDO-16620.[5] The nomenclature has been changed from that in the reference to the following:

$$\% \text{ Pu burnup} = \frac{(1 + \alpha)(Pu^{239}{}_b - Pu^{239}{}_a)}{Pu^{239}{}_b\,(1 + \alpha - Pu^{239}{}_a)} \times 100 \quad , \qquad (1)$$

where

$Pu^{239}{}_b$ = fraction of the plutonium which was Pu^{239} before irradiation

$Pu^{239}{}_a$ = fraction of the plutonium which was Pu^{239} after irradiation

$\alpha = \sigma_c/\sigma_f = 0.38$

and

σ_c = nonfission capture cross section (280×10^{-24} cm^2)

σ_f = fission cross section (746×10^{-24} cm^2).

From the burnup values obtained by this method, the average perturbed flux necessary to induce the burnups was computed by use of the equation

$$\text{Total a/o burnup} = Af\frac{\sigma_f}{\sigma_a}\left[1 - \exp\left(-\phi t \sigma_a\right)\right] \quad , \tag{2}$$

where

A = atomic per cent plutonium in fuel

f = fraction of the plutonium which is fissionable (0.95)

σ_a = absorption cross section (1026×10^{-24} cm^2)

ϕ = neutron flux (neutrons/cm^2-sec)

t = irradiation time (sec).

All values for nuclear cross sections were taken from reference 6.

From the results obtained with the 8 ANL-36 specimens, the average perturbed fluxes incident upon the other 22 specimens were obtained from the flux monitors. In addition, 13 of the ANL-36 specimens were in the same capsules and adjacent to specimens upon which isotopic analysis had been performed. Hence, knowledge of the flux incident upon these specimens was obtained. The fluxes obtained by these methods were used to compute the burnups and temperatures in the ANL-36 specimens not subjected to postirradiation isotopic analyses.

The burnups for the EA specimens were computed as follows. After irradiation, the oxidized material was removed by etching (in a mixture of 50 v/o H_2O and 50 v/o HNO_3) from the surface of 6 of the 7 specimens. Subsequently, a sample from the new clean surface of each of the 6 specimens was obtained (by etching), and these samples were analyzed mass spectrographically for plutonium isotope composition. The surface burnup of each specimen was then calculated by means of equation 1. The burnup value for the EA-7 specimen was obtained by interpolation from a flux and burnup plot of the 6 analyzed specimens. The average burnups in all of the specimens were obtained using the method of Taraba and Paine.(7) The preirradiation plutonium isotope composition and the post-irradiation surface plutonium isotope composition of the EA specimens are contained in Table IV.

Table IV

MASS-SPECTROGRAPHIC PLUTONIUM ISOTOPE COMPOSITION OF EA SPECIMENS

Specimen No.	Preirradiation Plutonium Composition, Mole Per Cent				Postirradiation Surface Plutonium Composition, Mole Per Cent			
	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}
EA-1	94.40 ± 0.05	5.21 ± 0.05	0.381 ± 0.004	0.0125 ± 0.0003	76.0 ± 0.2	20.0 ± 0.2	3.63 ± 0.03	0.35 ± 0.01
EA-2	94.40 ± 0.05	5.21 ± 0.05	0.381 ± 0.004	0.0125 ± 0.0003	78.0 ± 0.2	19.7 ± 0.2	2.10 ± 0.02	0.229 ± 0.002
EA-3	94.40 ± 0.05	5.21 ± 0.05	0.381 ± 0.004	0.0125 ± 0.0003	81.6 ± 0.1	16.6 ± 0.1	1.81 ± 0.02	0.0758 ± 0.0008
EA-4	94.40 ± 0.05	5.21 ± 0.05	0.381 ± 0.004	0.0125 ± 0.0003	83.7 ± 0.1	14.4 ± 0.1	1.68 ± 0.01	0.146 ± 0.001
EA-5	94.40 ± 0.05	5.21 ± 0.05	0.381 ± 0.004	0.0125 ± 0.0003	78.9 ± 0.2	18.1 ± 0.2	2.76 ± 0.03	0.262 ± 0.003
EA-6	94.40 ± 0.05	5.21 ± 0.05	0.381 ± 0.004	0.0125 ± 0.0003	73.0 ± 0.2	23.3 ± 0.2	3.35 ± 0.03	0.419 ± 0.004

The same properties which were measured prior to irradiation were measured after irradiation. In addition, a photograph of each specimen was obtained after irradiation.

IRRADIATION RESULTS AND DISCUSSION

Table V is a summation of the irradiation data obtained for the uranium-20 w/o plutonium-10 w/o fissium alloy, the reference fuel for EBR-II, Core II. The table is arranged in the order of increasing irradiation temperature. Figure 4 is a graphical representation of the rate of volume increase as a function of temperature for the reference fuel alloy specimens irradiated in the ANL-36 experiment. The data for the 13 specimens irradiated in the ANL-36 experiment fall within a band typical of that observed for other metallic fuels. The data for the 7 EA specimens do not display any systematic behavior, and the points were not utilized in constructing the swelling rate vs. temperature curves for the reference fuel alloy.

Table V

EFFECT OF IRRADIATION ON URANIUM-20 w/o PLUTONIUM-10 w/o FISSIUM FUEL ALLOY[a]

Specimen No.	Max Fuel Temp, °C		Burnup, a/o	Length Change, %	Diameter Change %	Density Decrease, %	Weight Change, mg	% ΔV [e] / a/o Burnup	Postirradiation Appearance
	Center	Surface							
ANL-36-2-2[b]	240	230	1.0	1.0	0.5	1.86	-65.7	3.9	Surface slightly roughened.
ANL-36-7-2[c]	290	270	1.2	1.1	0.3	3.21	-0.9	2.8	Good.
ANL-36-4-2	310	290	1.4	3.0	1.5	8.83	-9.1	6.9	Good.
ANL-36-4-3[d]	310	290	1.4	1.0	0.7	3.70	-40.5	2.7	Very good.
EA-3	320	240	2.3	(f)	7-17	36.2	-1.9	24	Broken in 2 pieces.
ANL-36-6-2	330	310	1.4	3.2	2.3	10.0	-6.0	7.9	Surface slightly roughened.
ANL-36-3-2	370	350	1.2	3.8	32	37.7	122.9	50	Top swollen and oxidized.
ANL-36-5-2	370	350	1.2	16	23	37.4	109.5	50	Bent 15°, top swollen.
ANL-36-5-3[d]	370	350	1.2	14	11	28.9	41.1	34	Top swollen.
ANL-36-9-1	370	350	1.7	1.8	4.3	9.82	-26.9	6.4	Surface slightly roughened.
ANL-36-9-3	370	350	1.7	3.2	1.9	11.9	-101.8	7.9	Good.
ANL-36-10-3	390	370	1.8	8.3	2.4	28.4	66.8	22	Bent 14°, surface roughened - radial cracks near top.
ANL-36-11-1	440	420	1.3	22	54	43.5	173.7	59	Bent 27°, swollen and cracked - oxidized looking.
ANL-36-8-3	470	440	1.3	18	32	47.4	156.3	69	Swollen, longitudinally cracked, top offset from bottom.
EA-1	510	370	3.0	(f)	20-26	39.4	-1.4	22	Bent 29°, top porous, cracked and oxidized.
EA-5	610	440	2.6	22	21-28	41.3	0.1	27	Bent 22°, top porous and oxidized.
EA-4	650	470	2.0	18-23	10-23	39.1	0.1	32	Broken in 2 pieces and bent.
EA-6	730	520	3.3	15	14-47	38.7	0.1	19	Bent 20°, top is cup shaped.
EA-2	800	530	2.7	~2.6	19-56	(f)	(f)	(f)	Bent 15°, top is slumped and porous.
EA-7	810	580	2.8	(f)	99	(f)	(f)	(f)	Unable to remove specimen from capsule.

(a) Specimens are injection cast except where otherwise indicated.
(b) Injection cast and annealed for 24 hr at 500°C.
(c) Pour cast and thermally cycled between 50°C and 500°C.
(d) Pour cast, extruded and thermally cycled between 50°C and 500°C
(e) Volume change obtained from the relation $\frac{V_{initial} - V_{final}}{V_{initial}} = 1 - \frac{\rho_{initial}}{\rho_{final}}$ and assuming no weight change (V = volume, ρ = density).
(f) Unable to obtain measurements because of the condition of the specimen.

The specimen which was heat treated in vacuum for 24 hr at 500°C and cooled in helium, and the pour-cast and thermally cycled specimens appear to be somewhat more stable under irradiation than the injection-cast material. The heat treatment or thermal cycling probably removed detrimental effects of the temperature gradient which was produced in the alloy during the injection-casting process.

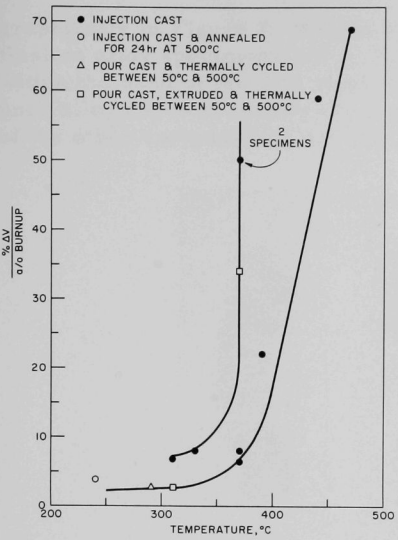
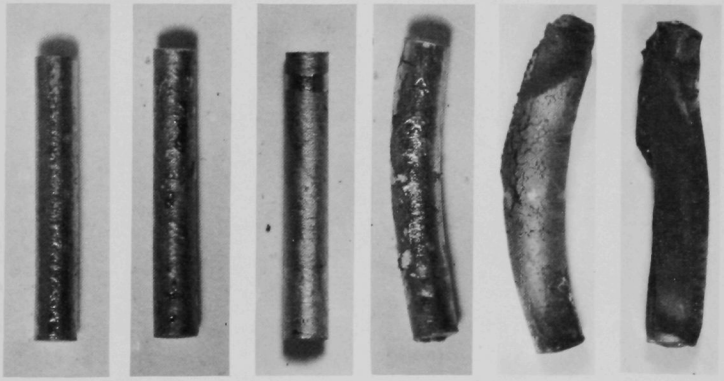


Figure 4. Effect of irradiation temperature on the rate of volume increase in the uranium-20 w/o plutonium-10 w/o fissium alloy specimens irradiated in the ANL-36 experiment.

The alloy swelled catastrophically above central fuel temperatures of approximately 370°C. Figure 5 shows the typical postirradiation appearance of reference fuel alloy specimens which were irradiated in the temperature range from 240 to 470°C in the ANL-36 experiment. Figure 6 shows the postirradiation appearance of 4 of the reference fuel alloy specimens irradiated in the EA experiment. All of the EA specimens showed most pronounced swelling at their upper ends. This behavior is believed to be the result of convection of the NaK in the capsules during irradiation, leading to excessive temperatures at the upper ends of the specimens.

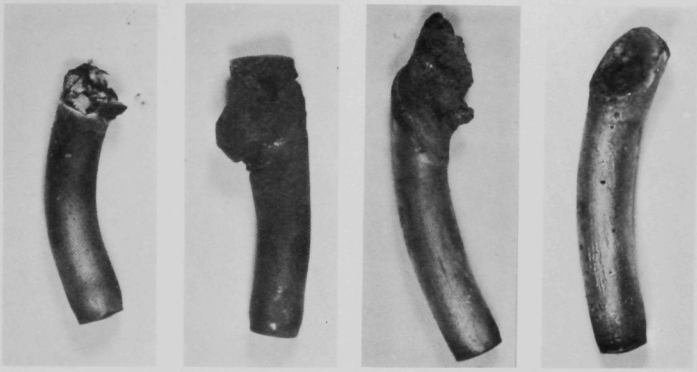
Table VI contains the irradiation information obtained from the other 17 specimens irradiated in the ANL-36 experiment. This information together with the average swelling curve for the reference fuel is shown



Neg. No.	27240	27246	27250	27253	25648	25644
Burnup, a/o	1.0	1.4	1.7	1.8	1.3	1.3
Max Fuel Temp, °C	240	330	370	390	440	470
%ΔV/a/o Burnup	3.9	7.9	7.9	22	59	69

Figure 5. Postirradiation appearance of typical uranium-20 w/o plutonium-10 w/o fissium fuel alloy specimens irradiated in the ANL-36 experiment. Magnification 2X.

graphically in Figure 7. Except for the 2 uranium-20 w/o plutonium-15 w/o fissium alloy specimens, all of the specimens had swelling rates which fall within the band of the data obtained for the reference fuel alloy. The reference alloy curve is included to enable one to compare the relative stability of the other compositions and fabrication histories which were investigated.



Neg. No.	23264	23266	23270	23271
Burnup, a/o	3.0	2.7	2.6	3.3
Max Fuel Temp, °C	510	800	610	730
% ΔV/a/o Burnup	22	Not Obtained	27	19

Figure 6. Postirradiation appearance of typical uranium-20 w/o plutonium-10 w/o fissium alloy specimens irradiated in the EA experiment. Magnification 2X.

Table VI

EFFECT OF IRRADIATION ON OTHER URANIUM-PLUTONIUM ALLOYS[a]

Specimen No.	Composition, w/o	Burnup, a/o	Max Temp, °C		Length Change, %	Diameter Change, %	Density Decrease, %	Weight Change, mg	% ΔV [e] / a/o Burnup	Postirradiation Appearance
			Center	Surface						
ANL-36-2-1	U-10 Pu-10 Fs	1.0	250	240	0	0.7	1.86	-0.4	2.0	Excellent.
ANL-36-3-1	U-10 Pu-10 Fs	1.1	370	350	1.7	1.6	4.95	-0.5	4.7	Good.
ANL-36-4-1	U-20 Pu-7 Fs-3 Zr	1.4	310	290	-1.2	2.6	8.81	-12.2	6.9	Good.
ANL-36-5-1	U-20 Pu-7 Fs-3 Zr	1.2	370	350	5.7	9.9	26.0	-5.1	30	Top began to swell.
ANL-36-6-1	U-20 Pu-10.5 Fs-4.5 Zr	1.4	330	310	3.7	3.0	11.8	-3.1	9.5	Good.
ANL-36-7-1	U-20 Pu-10.5 Fs-4.5 Zr	1.1	290	270	1.1	2.2	6.75	-1.4	6.5	Good.
ANL-36-6-3	U-20 Pu-15 Fs	1.4	330	310	5.9	4.6	16.4	21.2	15	Top distorted with longitudinal cracks.
ANL-36-7-3	U-20 Pu-15 Fs	1.1	290	270	6.1	18	17.1	39.1	20	Bent 15°, top end swollen and split open.
ANL-36-9-2	U-20 Pu-5 Fs[b]	1.7	370	350	3.4	5.3	20.8	-0.5	16	Top swollen.
ANL-36-11-2	U-20 Pu-5 Fs[b]	1.3	440	420	18	56	57.2	234.6	105	Bent 18°, top swollen, cracked, porous and oxidized.
ANL-36-10-1	U-20 Pu-5.4 Fs-3.6 Mo[c]	1.8	390	370	16	12	25.5	5.3	19	Bent 13°, top distorted.
ANL-36-10-2	U-20 Pu-5.4 Fs-3.6 Mo[b]	1.8	390	370	9.3	3.8	21.7	13.4	16	Top swollen.
ANL-36-11-3	U-20 Pu-5.4 Fs-3.6 Mo[b]	1.3	440	420	19	30	46.2	82.5	66	Bent 23°, top swollen, cracked, porous and oxidized.
ANL-36-8-1	U-20 Pu-5 Mo[c]	1.4	470	440	23	34	(d)	(d)	(d)	Entire specimen distorted and full of cracks.
ANL-36-8-2	U-20 Pu-5 Mo[b]	1.4	470	440	29	48	(d)	(d)	(d)	Entire specimen distorted and full of cracks.
ANL-36-2-3	U-25 Pu-10 Fs	1.1	260	240	1.2	-0.3	3.06	-52.9	2.8	Top half distorted.
ANL-36-3-3	U-25 Pu-10 Fs	1.2	370	350	21	34	33.4	87.4	42	Bent 30°, top swollen and began to crack.

(a) Specimens are injection cast except where otherwise indicated.
(b) Pour-cast, extruded and thermally cycled prior to irradiation.
(c) Pour-cast, and thermally cycled prior to irradiation.
(d) Specimen ignited and burned to oxide before measurements could be obtained.
(e) Volume change obtained from the relation $\frac{V_{initial} - V_{final}}{V_{initial}} = 1 - \frac{\rho_{initial}}{\rho_{final}}$ and assuming no weight change (V = volume, ρ = density).

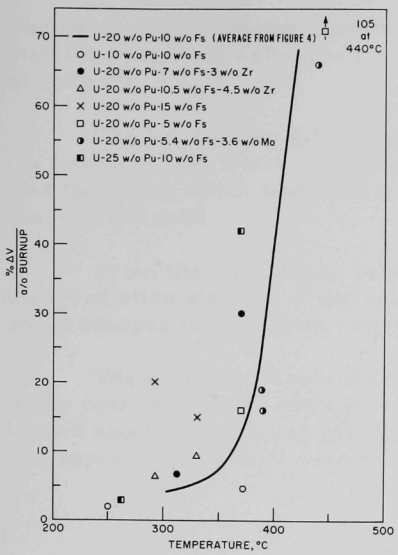
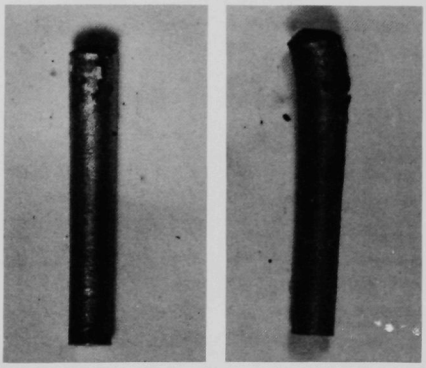


Figure 7. Effect of irradiation temperature on the rate of volume increase in several uranium-plutonium-fissium alloys.

As expected, the lower fissile atom density of the uranium-10 w/o plutonium-10 w/o fissium alloy resulted in this alloy exhibiting a higher degree of irradiation stability than the reference fuel.

The only material which had a swelling rate significantly different from that of the reference fuel was the uranium-20 w/o plutonium-15 w/o fissium alloy. In addition to having a larger swelling rate, the 2 specimens of this composition were cracked and distorted at the top ends, as shown in Figure 8. The lower irradiation stability of these specimens may be due to the presence of the intermetallic phase U_2Ru in the uranium-20 w/o plutonium-15 w/o fissium alloy; U_2Ru is not present in the alloys containing 10 w/o (or less) fissium.

Substitution of zirconium for fissium in the nominal ratio of one-half the fissium content to produce an alloy of uranium-20 w/o plutonium-10.5 w/o fissium-4.5 w/o zirconium increased the irradiation stability of this material to approximately that of the reference fuel.



Neg. No.	27247	25638
Burnup, a/o	1.4	1.1
Max Fuel Temp, °C	330	290
% ΔV/a/o Burnup	15	20

Figure 8. Postirradiation appearance of uranium-20 w/o plutonium-15 w/o fissium alloy specimens. Magnification 2X.

The additional zirconium combines with the ruthenium to form the intermetallic compound ZrRu; the formation of Zr Ru suppresses the formation of U_2Ru.(8)

The low fissium alloys which contain 10 w/o (or less) fissium and the fissium alloys which contain the additional zirconium are more ductile than the alloys which contain more than 10 w/o fissium (in which the U_2Ru phase is present).

From the information obtained in this experiment it appears that the irradiation stability of the reference fuel does not vary markedly with small changes in plutonium, zirconium, or fissium content.

The uranium-20 w/o plutonium-5 w/o molybdenum alloy had relatively poor irradiation stability. The 2 specimens of this alloy ignited and burned shortly after being photographed. Figure 9 shows the postirradiation appearance of these specimens.

Neg. No.	25642	25643
Burnup, a/o	1.4	1.4
Max Fuel Temp, °C	470	470

Figure 9. Postirradiation appearance of uranium-20 w/o plutonium-5 w/o molybdenum alloy specimens. Magnification 2X.

The poor behavior of the uranium-20 w/o plutonium-5 w/o molybdenum specimens and several of the other specimens irradiated in this experiment may not be entirely due to the swelling tendency of the material. As mentioned earlier, a vacuum of approximately 1×10^{-4} mm of Hg existed in the irradiation capsules prior to irradiation. Thermal expansion of the

NaK would raise the gas pressure to approximately 3×10^{-4} mm of Hg if one assumes a NaK temperature of 300°C and that the system exhibited ideal behavior. The vapor pressure of NaK at 300°C is 0.5 mm Hg. It is evident that under these conditions local or film boiling would occur. Under severe conditions this would lead to impaired heat transfer from the surface of the fuel, with resulting overheating of the specimen.

The only specimens in which catastrophic swelling has been observed at Harwell is reportedly in specimens which have been irradiated in capsules evacuated to low overpressures.(9) The catastrophic swelling is usually localized to certain areas in the specimen. This is thought to be caused by local film boiling which is likely to occur under conditions of low overpressure.

In the specimens irradiated at Argonne in capsules which have been evacuated to low overpressures, any catastrophic swelling usually occurs in the top half of the specimen, while the bottom half of the specimen undergoes normal swelling. In actual reactor operation the irradiation stability of these fuel alloys should be superior to that exhibited in this experiment since the fuel will not be subjected to the damaging effects of low overpressure above the liquid metal bond between the fuel and its jacket.

CONCLUSIONS

1. Uranium-20 w/o plutonium-10 w/o fissium alloy (the EBR-II Core-II reference fuel alloy) undergoes catastrophic swelling when irradiated at temperatures above 370°C.

2. The ability of the uranium-20 w/o plutonium-10 w/o fissium alloy to resist swelling does not vary appreciably with minor changes in zirconium or fissium content.

3. The swelling behavior of the alloy is significantly improved when the plutonium content is decreased to 10 w/o.

4. The uranium-20 w/o plutonium-5 w/o molybdenum alloy specimens had lower irradiation stability than did the other alloys investigated.

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